External Electric Field Mediated Quantum Phase Transitions in One-Dimensional Charge Ordered Insulators: A DMRG Study

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We perform density matrix renormalization group (DMRG) calculations extensively on one-dimensional chains with on site (U) as well as nearest neighbour (V) Coulomb repulsions. The calculations are carried out in full parameter space with explicit inclusion of the static bias and we compare the nature of spin density wave (SDW) and charge density wave (CDW) insulators under the influence of external electric field. We find that, although the SDW ($U > 2V$) and CDW ($U < 2V$) insulators enter into a conducting state after a certain threshold bias, CDW insulators require much higher bias than the SDW insulators for insulator-metal transition at zero temperature. We also find the CDW-SDW phase transition on application of external electric field. The bias required for the transitions in both cases decreases with increase in system size.

Strongly correlated electronic systems in low-dimension are always very interesting because of their unique characteristics like quantum fluctuations, lack of long range ordering etc [1, 2, 3, 4, 5, 6, 7]. The correlation effects in such low-dimensional systems lead to Mott or charge-ordered insulating states. The one-dimensional extended Hubbard model[8, 9] with on-site Hubbard repulsion term, U, along with the nearest-neighbour Coulomb repulsion term, V, is a standard model which exhibits these different phases. This is perhaps the simplest possible model which can capture many interesting properties of strongly correlated systems. In the strong coupling limit, this model gives rise to two insulating phases, spin-density-wave (SDW) and charge-density-wave (CDW) which are separated by a first order phase transition line at $U \simeq 2V$. On the other hand, in the weak coupling limit, the perturbative analysis shows that the transition is continuous at $U = 2V$[10, 11].

The effect of electric field on such insulating phases has attracted much interest in recent times due to the practical applications in tuning their dielectric and piezoelectric properties[12]. Many electronic conduction processes seem to suggest electric field induced phenomena, such as negative differential resistance in molecular electronics[13, 14]. Experiments on low-dimensional Mott-insulators with spin-density-waves and charge-ordered phases as ground states, suggest a collapse of insulating phase in presence of external electric field at finite temperatures [15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25]. However, the breakdown in charge ordered phases is not due to Joule heating. Rather it has been argued that, it is the applied bias which generates a conduction pathway resulting in metallic characteristics[18]. However, a tractable computational method which can take into account the static electric field and its response on the correlated extended electronic systems is still lacking.

In this article, we use the Density Matrix Renormalization Group (DMRG)[26, 27, 28] method which is known to be highly accurate for low-dimensional interacting systems with high precision. We have included electric field in the DMRG algorithm and have obtained ground and excited eigen states behaviors of one-dimensional CDW and SDW insulating systems with various system sizes. The static electric field is included as a ramp potential and we find that the electric field can induce an insulator-metal-insulator transition in both the cases, with a strong dependence on the Hamiltonian parameters. Instead of field per unit length, we consider the total field, applied between two ends of a one-dimensional chain, irrespective of the chain length. Similar consideration was applied for SDW insulators, where we extrapolated our finite size results to very large (effectively infinite) systems to obtain thermodynamic behavior[29]. In the present study on insulating CDW phase, we find the CDW-SDW quantum phase transition along with the breakdown of insulating phase on application of bias.

We consider one dimensional strongly correlated chain described by the extended-Hubbard Hamiltonian,

$$
\begin{aligned}
H = {} & t \sum_i (a^\dagger_i a_{i+1} + h.c) \\
& + U \sum_i n_{i\uparrow} n_{i\downarrow} + V \sum_i (n_i - n_{av})(n_{i+1} - n_{av})
\end{aligned}
\tag{1}
$$

where t is the hopping term, U is the onsite Hubbard repulsion term and V is the nearest-neighbour Coulomb repulsion term. We set $t = 1$ in all our calculations and express every energy in units of t. The $a^\dagger$ (a) is the creation (annihilation) operator and n (n_{av}) is the number operator (avarage electron density on every site). The external electric field applied on the system has the form of a ramp potential, distributed over all the sites in such a way that the potential F_i at site i becomes $-\frac{F}{2} + i\frac{F}{N+1}$, where F is the total applied bias and N is the total number of sites
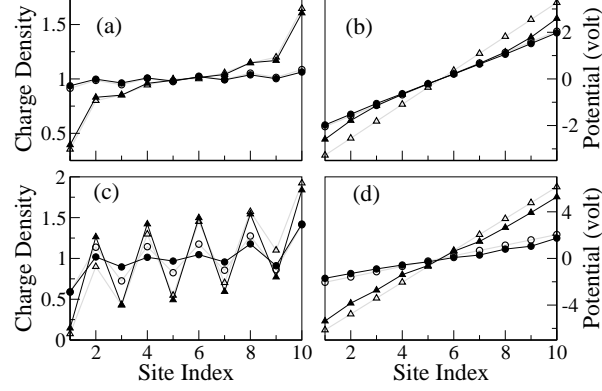
FIG. 1: The charge density (a) and the potential (b) on all the sites in SDW phase with $U = 5$, $V = 1$ and $F = 5(circle)$ and $8(triangle)$. (c) and (d) correspond to the charge density and potential respectively in CDW phase with $U = 5$, $V = 3$ and $F = 5(circle)$ and $15(triangle)$. Open and closed symbols correspond to calculations without and with self consistent Poisson's equation respectively for $N = 10$.

in the 1D chain. This form of the potential ensures that the bias varies between $-F/2$ to $F/2$ across the molecule. The potential adds an extra term $\sum_i F_i a^\dagger_i a_i$ to the above Hamiltonian. In DMRG, we use density-matrix cut-off, $m = 140$. For $U = 0$ and $V = 0$, the problem can be exactly solved and we obtain the ground state and excitation spectrum in presence of bias using the tight-binding one-electron formalism. We perform our calculations keeping a fixed value of U $(= 5)$ and varying the V from 0 to 4 with bias from 0 to 10 volts in steps of 0.5 volts.

With increase in the value of V, one encounters three different regions, namely, spin-density wave $(2V < U)$, spin-density-wave to charge density wave crossover $(2V = U)$ and charge-density-wave $(2V > U)$. To understand the effect of electric field on the excited states of the system, for a given value of U and V, we have computed the energies of the systems with one extra $(E(N + 1))$ and one less electron $(E(N - 1))$ than half-filling $(E(N))$ and have calculated the many body charge excitation gap as the difference between the energy required to add (μ_+) and remove (μ_-) electrons from the ground state[30],

$$\Delta_{charge} = \mu_+ - \mu_- \qquad (2)$$

where $\mu_+ = E(N + 1) - E(N)$ and $\mu_- = E(N) - E(N - 1)$.

To investigate the effect of polarization on the applied electric field, i.e., the screening of external electric field by the shifted electron density, we solve the self consistent Poisson's equation in SDW as well as in CDW phases. We have considered the boundary conditions so that the final electrostatic potential field extending from one electrode to another becomes different for different atomic sites and the atomic electron density will adjust to the field in such a way that the charges are stabilized locally. We start our self consistent calculation by assuming that the electrostatic field is a linear ramp function across the interface of electrode and the system. By solving the Schrodinger equation within many-body limit we obtain the charge density at every site and use that as an input in one-dimensional Poisson's equation. The diagonal elements of the Hamiltonian is then modified with the modified bias, obtained from Poisson's equation. Solving the modified Hamiltonian we obtain the charge densities and again use them as input in Poisson's equation. This is continued untill all the charge densities and all the site potential fields converge. In Fig.1 we plot both the on-site atomic charge densities and the spatial distribution of potentials for both SDW and CDW phases with explicit consideration of the screening of applied electric field and show the same, without considering the screening, for comparison. From Fig.1 it is clear that, in the presence of strong electronic correlations, the ramp nature of the external electric field is retained without any significant change of the charge density or potential profile with the consideration of polarization effect. Here we consider the system with $N = 10$ and use exact diagonalization method and present the results for two different bias values. The ground state of the CDW phase is a linear combination of two configurations (having alternating doubly occupied sites) with same weightage. So we take the average of the charge densities and the potentials of the two degenerate ground states in case of CDW phase. Moreover, for CDW phase we consider fairly large value of bias to investigate the breakdown region, which we discuss later. To infer, large correlations reduces the Poisson's equation into Laplace equation with no significant variation of the charge
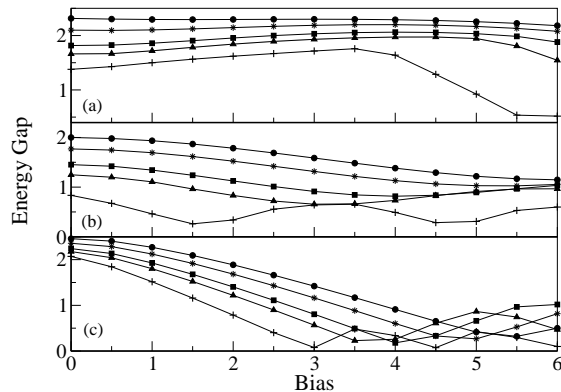
FIG. 2: Variation of charge gap as a function of bias for (a) CDW ($V = 3$), (b) SDW-CDW crossover ($V = 2.5$) and (c) SDW ($V = 1$) insulators, for various systems sizes (N): $N = 10(circle), 12(star), 16(square), 20(triangle)$ and $40(plus)$.

density distribution which we observed in our earlier studies also[31, 32]. The inclusion of the effect of polarization can change the quantitative estimation of the results without any significant change of the physics. So, to avoid the computational expense of the self consistent Poisson's equation calculations, we ignore the polarization effect in our further DMRG calculations with larger system sizes.

In Fig.2(a), (b), and (c) we have plotted the charge gap of 1-D chains with different N as a function of bias for CDW, SDW-CDW crossover region and SDW phases, respectively. For SDW phase and SDW-CDW crossover region, the charge gap decreases with increasing bias upto a minimum, whereas in case of CDW phase, the charge gap increases initially with increasing bias. In the latter case, the nearest-neighbour Coulomb repulsion term, V, is higher than the previous two cases, and as a consequence, the electrons favour to remain paired in alternative sites than increasing its kinetic energy by hopping to the neighboring sites. The SDW phase has a charge gap of U at $U \to \infty$ limit. But the CDW phase with double occupancy at alternative sites for a half-filled 1-D chain has smaller charge gap than U and at $U, V \to \infty$ limit, the charge gap is $(U - V)$. Therefore, in the CDW phase, the V is the stabilizing term and it can be proved unambiguously from the model Hamiltonian. Bias, however, tries to make the electrons hop in its direction and as a consequence the site occupancies start changing, leading to a higher charge gap. It is thus the hopping, which creates larger gap in charge-ordered phase inducing positive contribution from V. However, this initial increase is followed by a decrease in charge gap at quite a higher bias, although, the charge gap minimum can not be seen for all the system sizes in the bias window, we have considered. In case of SDW phase, the bias overcomes the effect of U by forcing the electrons to hop in its direction and leads the system to a charge gap minima corressponding to a conducting state. So, it is clear from Fig.2 that, the CDW phase takes much higher bias to pass through a charge gap minimum than the SDW phase. The SDW-CDW crossover region shows combination of the properties of both SDW and CDW phases. It can also be seen from Fig.2 that, the increase in system size reduces the initial charge gap for all the three cases and reduces the bias corressponding to the first charge gap minimum in case of SDW phase and SDW-CDW crossover region.

To understand the response of the ground state as well as the excited states under the influence of external electric field, we plot $E(N + 1)$, $E(N)$ and $E(N - 1)$ of a system with 40 sites as a function of bias in Fig.3 for CDW phase ($V = 3$), SDW-CDW crossover region ($V = 2.5$) and for SDW phase ($V = 1$). For all the three cases, the ground state energies do not change much upto a threshold bias. However, beyond that, the systems start to stabilize. The slope of the ground state energy is different from those of excited states for all the three phases and this difference in slope gives rise to collapse of insulating phase at some threshold bias, which is different for different phases and strongly depends on the system size. In contrast to the first two cases, although the CDW phase with one less electron starts getting stabilized with increasing bias, the system with one more electron destabilizes slightly in low bias followed by lowering in energy in higher bias values. It is due to the fact that, one more electron in CDW phase experiences more prominent elctronic repulsion for bias driven hopping because of higher V value. As a consequence, the charge gap increases initially, however, beyond a certain bias value, required to nullify the effect of V, the gap reduces.

For a clear understanding of the charge gap variation in CDW insulating phase with increasing bias, in Fig.4 we plot the charge gap, Δ_{charge} as a function of bias for $V = 3$, for finite systems with $N = 20$ and $N = 40$ sites. As can be seen clearly, the charge gap increases initially upto a certain bias and then it starts decreasing approching a minimum
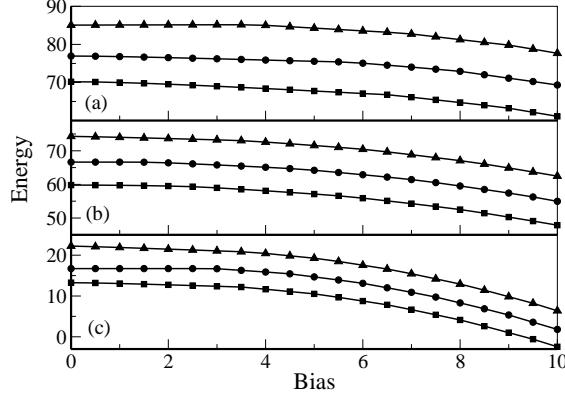
FIG. 3: Energies, $E(N+1)$ (*triangle*), $E(N)$ (*circle*) and $E(N-1)$ (*square*) as a function of bias for (a) $V = 3$, (b) $V = 2.5$ and (c) $V = 1$, for the system with 40 sites.
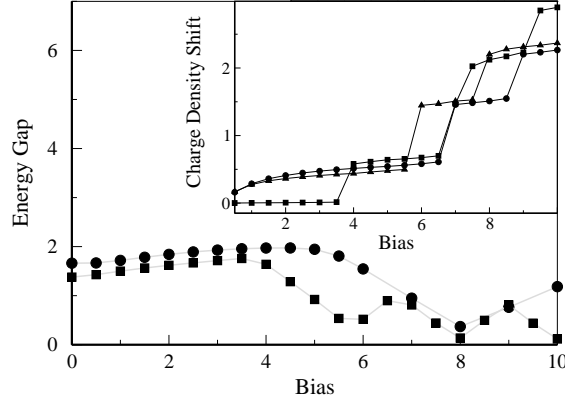

FIG. 4: Charge gap as a function of bias for system sizes, $N = 20$ (*circle*) and $N = 40$ (*square*) for $V = 3$. Inset shows the average charge density shift per site as a function of bias for $N + 1$ (*square*), N (*triangle*) and $N - 1$ (*circle*) electrons for system size $N = 40$.

at some threshold bias. Further increase in bias drives the system to go through several minima. On the contrary, the charge gap in SDW phase starts decreasing with even small nonzero bias and it goes through several minima with increasing bias as evident from Fig.2 as well as in our previous study[29]. In CDW phase, the initial increase in charge gap is only due to the nearest neighbour Coulombic repulsion term which prevents the hopping of electrons. After a certain bias, as depicted in the Fig.4, the charge gap starts decreasing and then onwards the variation of charge gap with increasing bias looks very similar as found in the SDW phase. From this, we can anticipate qualitatively that, on application of external bias, the CDW phase undergoes a phase transition at some bias, V_{trans}, and beyond that starts to behave like a SDW phase. At higher bias values, the periodic oscillation of charge gap in charge ordered phase makes the system to go through several insulator-metal-insulator transitions. However, from Fig.4 it is clear that, the period of oscillation becomes narrower with increase in the size of the system as observed in our previous study[29] on SDW phase.

To understand the underlying reasons for such behaviour of the charge gap, in the inset of Fig.4, we plot average charge density shift per site for the $N = 40$ one-dimensional chain with $N + 1$, N and $N - 1$ electrons as a function of bias. As can be seen, although all the three states show staircase like behavior, for the system with one extra electron, the nature of the shift of the charge densities is completely different from the other two cases. The charge density shifts slowly at lower bias in case of half-filled system and the system with one less electron, whereas the charge density shift for the system with one extra electron is almost zero upto a certain bias, termed as V_{trans}, for
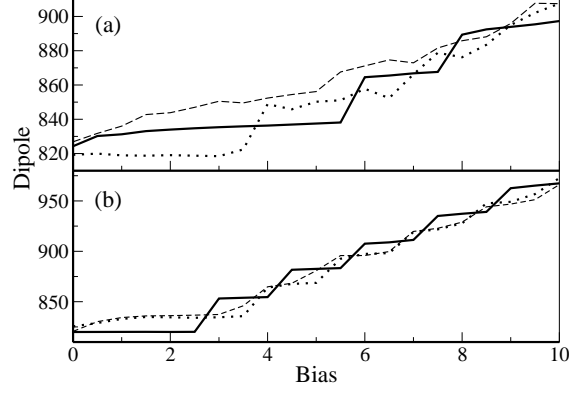
FIG. 5: The dipole moment as a function of bias for (a) $V = 3$ and (b) $V = 1$ for $N + 1 (dotted\ lines)$, $N (solid\ lines)$ and $N - 1 (dashed\ lines)$ electron states for a system size of $N = 40$.

this system, beyond which, the charge density shows a drastic shift. It is due to the fact that, the extra electron faces more prominent effect of Coulomb repulsion terms and hopping of electrons is possible only at some bias value which can overcome the electronic repulsion. After V_{trans}, the CDW phase again goes to an insulating phase, characterized by the plateau in the plot, which we anticipate to be a SDW phase. If we further increase the bias, after certain values, the shift in charge densities for all the three systems show sudden jump, which indicates kinetic stabilization, followed by plateaus, indicating reappearance of insulating phase. We argue that this repetetive period of charge gap with increase in bias is due to the charge stiffness. The external bias sweeps the charge densities towards one direction with nullification of repulsion terms upto the first minima. Beyond this, the bias tries to shift the charge densities further and the electronic repulsion terms again reappear and start opposing the kinetic stabilization, and as a consequence, the charge gap again increases and the system enters into an insulating phase. Thus, this repetetive metallic and insulating behaviour in different bias regions is a consequence of the interplay between the external bias and the electron-electron interaction terms. However, this oscillation of the charge gap with increase in bias is due to finite size effects. With increase in system size, the periodicity narrows down and we can anticipate that, at infinite system size, all the minima will converge to the same bias value as evident from our previous study[29]. We termed this bias value at thermodynamic limit as the critical bias, V_c. Note that, as we discussed earlier, we do not consider the effect of polarization on applied electric field in our calculations. It can only change the quantitative estimation of V_{trans} and the critical bias, V_c without any qualitative change in the basic physics.

To obtain a better insight, we plot the dipole moment of the ground state and the states with one extra and one less electron for both SDW and CDW phases as a function of bias in Fig.5. It can be seen clearly that, the ground state dipole moment shows periodic jumps at the bias values, corresponding to the charge gap minima and the plateaus indicate the successive insulating phases. So, whenever there is a shifting of charge densities i.e., sudden increase in the dipole moment, the system allows the electrons to hop, leading to breakdown of insulating phase. This periodic jumps of the dipole moment is purely due to finite size effects. However, the CDW-SDW phase transition can not be understood from the ground state dipole moments.

To understand the CDW-SDW phase transition, we calculate the order parameter[10] in terms of onsite charge densities ($\langle n_i \rangle$) for a finite system, consisting of 40 sites with half-filling and with one less and one more electron than half-filling.

$$m = \frac{1}{N} \sum_i (-1)^i \langle n_i \rangle \qquad (3)$$

and plot its absolute value in Fig.6. As can be seen, after a certain bias, the $|m|$ value for the system with one extra electron drops, indicating clearly a phase transition to SDW phase. The $|m|$ values for the other two cases also show drops at some bias values, which indicates the insulator to metal phase transition. From the above observations, we can conclude that, the system with one extra electron can directly indicate the CDW-SDW phase transition on application of external bias. Although the $|m|$ values show oscillations in Fig.6, we can interpret them as purely finite size effect, as described earlier. From these observations, we adequately conclude that, the extarpolation of our results
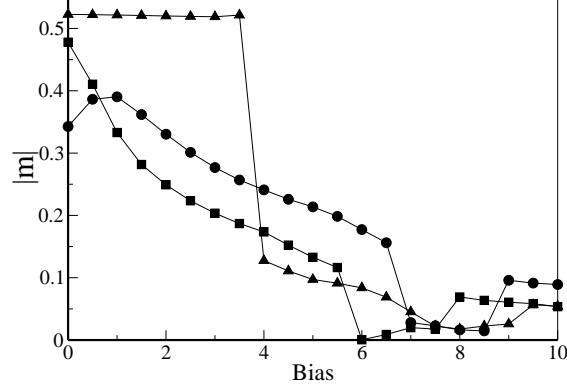
FIG. 6: Absolute value of the order parameter, m for CDW phase ($V = 3$) with $N + 1$ (*triangle*), N (*square*) and $N - 1$ (*circle*) electrons as a function of bias.
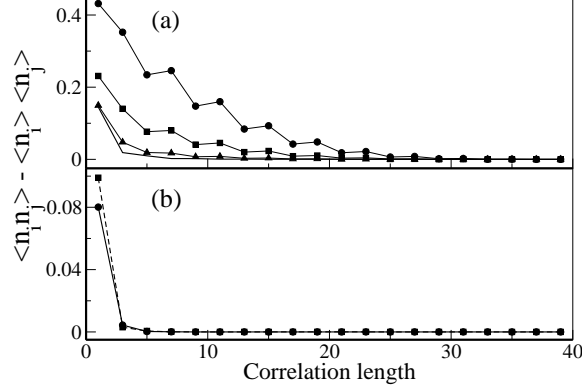

FIG. 7: The charge-charge correlation as a function of correlation length for (a) $V = 3$ at bias values 0.0(*circle*), 0.5(*square*), 1.0(*triangle*) and 5.5(*solid line without symbol*) volts and for (b) $V = 1$ at bias values 0.0(*circle*) and 6.0(*square*) volts for the system with 40 sites.

to infinite chain length will result into a single drop in the order parameter and take the CDW phase to a SDW phase, through the quantum phase transition at thermodynamic limit.

However, under the influence of external electric field which sweeps the electrons in its direction, the spatial charge distribution becomes nonuniform, resulting in mixture of locally confined domains of CDW and SDW orders. As a consequence, the spatial charge distribution becomes less significant to be considered as an order parameter. Instead, the charge-charge correlation function ($\langle n_i n_j \rangle - \langle n_i \rangle \langle n_j \rangle$) becomes more informative to characterize the phases. In Fig.7, we have shown the ground state charge-charge correlation as a function of distance for both CDW and SDW phases for a system with 40 sites. The nature of the zero-bias correlation is completely different in these two cases. In case of CDW phase, the correlation is known to be long range, whereas for SDW phase it is short ranged. Fig.7(a) clearly shows that, the correlation length for CDW phase decreases with the increase in bias and after a certain bias it behaves exactly like the SDW phase, depicted in Fig.7(b). This resemblance in the behavior of correlation function strongly indicates the CDW-SDW phase transition.

In conclusion, we have studied the application of external electric field on both the SDW and CDW insulating phases with on-site and nearest-neighbour Coulombic repulsion. We find that the external electric field can induce a insulator-metal transition in both the cases. Moreover, the electric field induces a CDW-SDW quantum phase transition as well. Increase in system size increases the sharpness of both the transitions. Our DMRG calculations allow us to study large systems with explicit application of electric field, with microscopic understanding of the

insulator-metal transition in spin-density-wave and charge-density-wave insulators.

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